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Morphology and photoelectronic properties of the InAs/GaAs surface quantum dots grown by vapor phase epitaxy

I. A. Karpovich, N. V. Baidus, B. N. Zvonkov, S. V. Morozov,
D. O. Filatov and A. V. Zdoroveishev

University of Nizhny Novgorod, Nizhny Novgorod 603 950, Russia

Abstract. A comparative study of the surface morphology, photoluminescence and photoelectric properties of the heterostructures with InAs/GaAs quantum dots (QDs) grown on the surface and uncovered by etching away the GaAs cladding layer has been carried out. The red shift of the ground transition energy in the surface QDs compared to the QDs embedded into the GaAs matrix has been shown to be related not only to relaxation of the elastic strain but also to the differences in the size and shape of the nanoclusters.

Investigation of the surface quantum dots (SQDs) are interesting because of possibility to establish a direct correlation between their photoelectronic properties and morphology determined by Scanning Probe Microscopy. SQDs differ from the QDs built in a wider gap semiconductor matrix (buried QDs or BQDs) by different field of the elastic strain and another shape of the potential well that effect on their energy spectrum essentially. Surface location of the SQDs allows to apply various processings (oxidation, etching, chemical modification, etc.). On the other hand, the photoelectronic properties of the SQDs are difficult for investigation due to a number of reasons, so far they are much less investigated to date than the BQDs [1, 2]. In the present work the morphology, photoluminescence (PL), and photovoltage spectra at the semiconductor/electrolyte interface (PSE) of the InAs/GaAs SQDs and BQDs grown by Atmospheric Pressure Metal-Organic Vapor Phase Epitaxy (AP-MOVPE) [3, 4] have been studied. It is known that deposition of a cladding layer may influence on the QDs formation essentially especially during vapor-phase epitaxy [5]. Using preferential chemical etching for removing the cladding layer [3] one can turn the BQDs into SQDs (further referred to as etched SQDs or ESQDs in order to distinguish between ones and as-grown SQDs). In this work the morphology of the ESQDs was also studied.

In Figures 1(a) and 1(b) the Atomic Force Microscopy (AFM) images of the SQDs and ESQDs respectively are shown. The typical lateral base size of the SQDs was ≈ 40 nm and height ≈ 6 nm. The ESQDs usually have less height (≈ 4 nm) but larger base size (≈ 50 nm). Their surface density was as a rule 1.5–2 times higher than the one of the SQDs

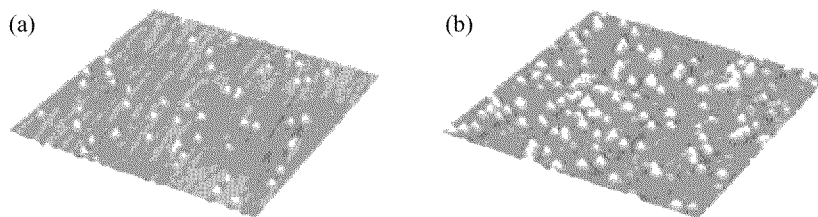


Fig. 1. AFM images of the SQDs (a) and ESQDs (b). Frame size: 1000×1000 nm; vertical scale: 6 nm.

$((4-9) \times 10^9 \text{ cm}^{-2})$ that was attributed to suppression of coalescence of the nanoclusters by deposition of the cladding layer.

The difference in sizes between the SQDs and the ESQDs can be explained as follows. In [3] we have suggested formation of a graded composition $\text{In}_x\text{Ga}_{1-x}\text{As}$ transition layer at the QD's heterointerface because of diffusion intermixing of In and Ga during overgrowth of the BQDs by GaAs cladding layer. This effect should increase the volume where the In atoms are distributed around a BQD as compared to the volume of a SQD. Since the etchant used for removing the cladding layer has high selectivity, the layer where In fraction exceeds some threshold value x should be a stop-layer for etching process (the more selectivity, the lower the threshold x). So far, as the surface $x = \text{const}$ determines the topography of the ESQDs, the latter should be larger in size than the SQDs. Otherwise, if no In redistribution during overgrowth takes place the etched surface should represent a sharp InAs/GaAs heterointerface.

PL measurements in the (E)SQDs are rather, more difficult because of a channel of the surface non-radiative recombination which decreases the PL intensity in 3–4 orders of magnitude as compared to the BQDs PL. While the latter can be observed easily even at room temperature the PL from the (E)SQDs can be observed at 77 K only. Typical PL spectra (77 K) from the SQDs and BQDs obtained at the same growth conditions are presented in Fig. 2. The PL peak from the SQDs is shifted by 350–400 meV to lower energies (curves 1 and 5), the lowest peak maximum position observed was 0.72 eV, which corresponds to the emission wavelength $1.72 \mu\text{m}$. The fact that this peak is from the SQDs was proven by etching away the surface oxide layer in HF. After formation and etching of the oxide has been observed a blue shift of the PL peak from the SQDs by 80 meV (curves 1–3) which was explained by decreasing of the volume of the SQDs. The spectral position of the higher energy shoulder of the peak at 0.85 eV remains the same. This shoulder should be obviously attributed rather to some deep levels in the epilayer than to the SQDs.

The red shift of the PL peak takes place also at formation of the ESQDs but its value is much less ($\approx 150 \text{ meV}$) than in the SQD heterostructures (curves 4 and 5). This indicates that the change in the field of the elastic strain is not the only cause for the shift of the transition energies. The differences between the SQDs and BQDs in size, in shape, and in the chemical composition arising during overgrowth probably also introduce comparable

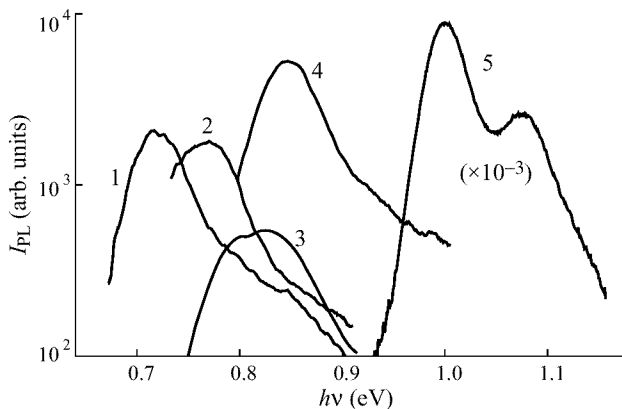


Fig. 2. Photoluminescence spectra of the quantum dots: SQDs (1–3), ESQDs (4), and BQDs (5). Curves 2 and 3 were obtained after first and second removing of the oxide layer in HF respectively.

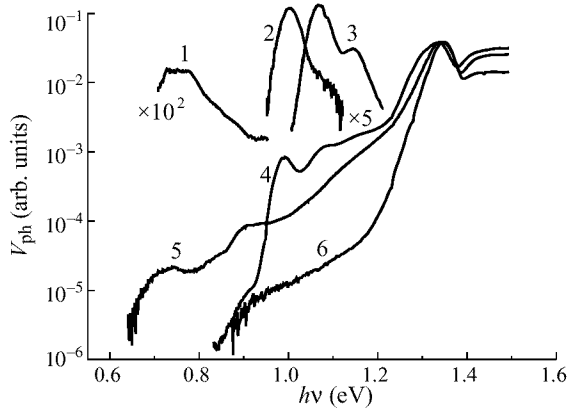


Fig. 3. PL (1–3) and PSE (4–6) spectra of the QD heterostructures: 2–4 — BQDs; 1, 5 — SQDs; 6 — GaAs. 2, 4–6 — 300 K; 1, 3 — 77 K.

contributions in the red shift.

In difference from the PL measurements the main problem in the photoelectric measurements in the SQDs is related not to the surface recombination but to low values of the ground transition energy in the SQDs. Increasing of the barrier height decreases the probability both of direct and thermally activated tunneling emission of the electrons from the SQDs into the matrix and, in turn, the photosensitivity. High sensitivity of the PSE method allowed to measure the photoelectric spectra from the SQDs at its surface density $\sim 10^{10} \text{ cm}^{-2}$.

In Figure 3 the PSE spectra of the 3 structures grown at the same conditions: BQDs (curve 4), SQDs (curve 5), and a homogeneous GaAs layer similar to the buffer in the QD structures (curve 6) are presented. A broad and weak peak of the SQDs photosensitivity at $\approx 0.7 \text{ eV}$ is shifted to lower energies by $\approx 0.3 \text{ eV}$ from the BQDs peak and is approximately in 2 orders of magnitude weaker than the latter at close values of the QD surface density, in correlation with the PL spectra of these samples (curves 1, 3). This peak crowns the wide band of approximately exponential decay of the photosensitivity within the band $0.75\text{--}1.2 \text{ eV}$ which was attributed to the surface states (SSs) below the Fermi level at the surface having quasi-continuous energy distribution. Similar band can be observed in the photosensitivity spectra of the GaAs homogenous epilayers (curve 6) and bulk single crystals as well. It is interesting that on the GaAs surface covered by InAs wetting layer, which is obviously oxidized fully between the SQD clusters, the SS density is much higher (curve 5) than on the surface covered by a natural oxide in the BQD structure and in the homogeneous layer.

In the photoelectric spectra of all GaAs based structures, homogeneous GaAs layers and GaAs single crystals a more or less expressed photosensitivity edge at $\approx 0.9 \text{ eV}$ has been observed (Fig. 3) which can be related to some single-level surface states in the native oxide on the GaAs surface. The density of these states also increases on the surface covered by a SQD layer.

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